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Review of recent advances in anaerobic packed-bed biogas reactors

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ABSTRACT

Biogas digesters have captured many imaginations because they can turn organic wastes from our farms, factories and cities into a valuable source of renewable energy. In addition, the potential of this technology to reduce odors and other environmental concerns of animal feedlots has resulted in much recent interest from researchers and scientists all over the globe.

Despite its numerous advantages, the potential of biogas technology could not be fully harnessed or tapped, as certain constraints are also associated with it. Researchers have tried different techniques to enhance gas production. This paper reviews the use of fixed-bed reactors in various fields reported by different researchers using variety of substrates.

Contents

1. Introduction

Energy sources can broadly be classified into two categories; namely non-renewable and renewable sources. In developing countries, there has been an increased interest in the development of technologies for harnessing renewable energy sources such as biomass either directly or through conversion routes. One of the biological processes, i.e. anaerobic digestion, has received a new fillip in recent years since the energy crisis of the early 1970s, especially following the Gulf war [1]. The process involves the treatment of agricultural and industrial waste of varying types in the production of biogas. Biogas is lighter than air and can accumulate under roofs and ceilings and create a fire or explosion hazard.

Interest in the anaerobic treatment of agro-industry waste is increasing because it is economical, has lower energy requirements and is ecologically sound, among several other advantages, compared with aerobic treatment processes [2,3]. The process produces digested sludge, which is mainly used as fertilizer for crop production since the nutrients in the raw material remain in the mineralized sludge as accessible compounds [4]. Treating waste to yield fuel while recycling nutrients constitutes a sustainable cycle.

Anaerobic digestion is a complex, natural, two-stage process of degradation of organic compounds through a variety of intermediates into methane and carbon dioxide, by the action of a

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consortium of microorganisms [5,6]. The interdependence of the bacteria is a key factor in the anaerobic digestion process. In the first stage, the volatile solids in manure are converted into fatty acids by anaerobic bacteria known as "acid formers." In the second stage, these acids are further converted into biogas by more specialized bacteria known as "methane formers." With proper planning and design, this anaerobic digestion process, which has been at work in nature for millions of years, can be managed to convert a farmer's often problematic waste-stream into an asset [7].

Instability during both the start-up and operation of the anaerobic degradation process can be problematic due to the low specific growth rate of the methanogenic microorganisms involved [8]. Under conditions of unstable operation, intermediates such as VFAs and alcohols accumulate at different rates depending on the cause of the instability [5]. The most common causes of imbalance are hydraulic or organic overloading, the presence of toxins and changes in the substrate concentration. Several parameters are used as indicators of stress, such as variations in gas production rate, gas composition, pH, PA and VFA concentration [9–11].

2. Overview of various digesters

Anaerobic digestion process takes place in a warmed, sealed airless container (*the digester*) which creates the ideal conditions for the bacteria to ferment the organic material in oxygen-free conditions. The digestion tank needs to be warmed and mixed thoroughly to create the ideal conditions for the bacteria to convert organic matter into biogas (a mixture of carbon dioxide, methane and small amounts of other gases). The proper design and scale-up of anaerobic reactors requires knowledge regarding correlations between reactor configuration and process efficiency to set the limits for the biochemical operating parameters.

There are two types of AD process:

- *Mesophilic digestion*. The digester is heated to 30–35 °C and the feedstock remains in the digester typically for 15–30 days. Mesophilic digestion tends to be more robust and tolerant than the thermophilic process, but gas production is less, larger digestion tanks are required and sanitisation, if required, is a separate process stage.
- *Thermophilic digestion*. The digester is heated to 55 °C and the residence time is typically 12–14 days. Thermophilic digestion systems offer higher methane production, faster throughput, better pathogen and virus 'kill', but require more expensive technology, greater energy input and a higher degree of operation and monitoring. During this process 30–60% of the digestible solids are converted into biogas. Various Biogas Digesters are described as follows.

2.1. Complete mix digesters

Complete mixed digester vessels are insulated and maintained at a constant elevated temperature, in the mesophilic or thermophilic range. The digester vessel is usually a round insulated tank, above or below ground and made from reinforced concrete, steel or fiberglass. They can be mixed with a motor-driven mixer, a liquid recirculation pump or by using compressed biogas. A gas tight cover (floating or fixed) traps the biogas [12].

2.2. Plug flow

A plug flow digester vessel is a long narrow (typically a 5:1 ratio; five times as long as the width) insulated and heated tank made of reinforced concrete, steel or fiberglass with a gas tight cover to capture the biogas. These digesters can operate at a mesophilic or thermophilic temperature. The plug flow digester has no internal agitation and is loaded with thick manure of 11–14% total solids. This type of digester works well with a scrape manure management system with little bedding and no sand. Retention time is usually 15–20 days [12].

2.3. Covered lagoons

A covered lagoon digester is a large anaerobic lagoon (not a manure storage pond or basin) with a long retention time and a high dilution factor. Typically covered lagoons are used with flush manure management systems that discharge manure at 0.5–2% solids. The in-ground, earth or lined lagoon is covered with a flexible or floating gas tight cover. They are not heated and considered ambient temperature digesters. Retention time is usually 30–45 days or longer depending on lagoon size [12].

2.4. Fixed film

A fixed film digester vessel is filled with an inert medium or packing that provides a very large surface area for microbial growth. The influent passes through the media and anaerobic microbes attach themselves to it creating a thin layer of anaerobic bacteria called biofilm—this film gives the digester its name, fixed film. These microbes then continue to grow by removing material from the wastewater as it flows by. In most digesters the microbes are floating in the liquid and a portion of these active growing workers are continuously discharged with the effluent. In a fixed film digester the bacteria remain attached to the plastic media when effluent is discharged. The "bacteria are already at work" when new influent is added [13]. Fixed film digesters have smaller reactor vessels, shorter retention times and must be loaded with a feedstock that will readily flow through the media without clogging. Three to five day retention times are typical and digesters can be run at ambient temperatures in hot climates but are usually heated to mesophilic or thermophilic temperatures [12].

3. Advantages of anaerobic packed-bed reactor

Increased stability and performance in anaerobic reactors can be achieved if the microbial consortium is retained in the reactor. Two means of achieving this are to use dense bacterial granula as in UASB reactors or a microbial biofilm attached to inert carriers in packed-bed reactors. The packing medium in the packed-bed reactor and the granular sludge in the UASB reactor serve as a filter preventing bacterial washout and also providing a larger surface area for faster biofilm development and improved methanogenesis [14,15]. Specific surface area, porosity, surface roughness, pore size, and orientation of the packing material were found to play an important role in anaerobic reactor performance [16,17]. Biofilm or fixed-film reactors depend on the natural tendency of mixed microbial populations to adsorb onto surfaces and to form a biofilm. Many carrier materials have been investigated regarding their suitability as supports for biofilm, including cheap, readily available materials like sand, clay, glass, quartz and a number of plastics [8,18,19]. In nature, microorganisms inhabit the outer and inner surfaces of stone, gravel or sand. This biofilm formation becomes an important factor for water self-cleaning ability. The growth of microorganism in a biofilm is the basis for biological water treatment such as denitrification and for intensification of aerobic and anaerobic wastewater treatment. The use of packed-bed reactors to treat different kinds of wastewater has also been reported, for example, dairy and brewery wastewater [18]. The biofilm formation on carrier materials improves the conversion

rates by reducing its sensitivity toward concentration variations and inhibiting substance. Picanco et al. [20] reported that the efficiency of removing organic matter in fixed-bed reactors is directly related to the characteristics of the support material used for immobilization of anaerobes.

It is widely accepted that organic support material has a higher affinity than inorganic material [21]. Reticular polyurethane foam has a high specific surface area which can reach 2400 m^2/m^3, and a porosity of 97%. It appeared to be an excellent colonization matrix for an anaerobic filter reactor [22]. Pore size was one of the most important parameter for microbiological and engineering requirements in high-efficiency beds [23]. Many kinds of bedding model have been considered for degrading a variety of organic wastes in anaerobic digestion reactors. A simple, low-cost and high-efficiency bedding method is necessary for practical use.

The development of fixed biomass reactors has ensured that significant advances in the knowledge and application of anaerobic processes for waste treatment have taken place. Compared to conventional units, fixed film bioreactors perform efficiently at higher organic loading rates (OLR), due to more effective biomass retention in the reaction zone resulting in higher cellular retention times. Immobilized biomass anaerobic reactors also show better responses to organic shock loads and toxic inputs. In many cases, immobilized biomass reactors completely recover their performance after such deleterious occurrences [24,25].

4. Innovations in PBR in various fields

Fixed film reactors have been used since long for the treatment of wastewater where they have helped to reduce the hydraulic retention times (HRT) from 30–40 days to a few hours [26]. These reactors come under the category of advanced reactors like UASB, fluidized bed, upflow anaerobic filters, etc. They help in enhancing the performance of wastewater treatment systems by providing an increased surface area for attached growth of the microbes in the form of a fixed film on an inert medium leading to increased population of microbes in the reactor and their retention in the digester even after the digested slurry flows out [27]. In packed-bed reactors (sometimes called fixed-bed reactors) wastewater is passed in either upflow or downflow mode over a population of microorganisms attached to an inert solid support carrier, e.g., gravel, plastic carriers, ceramic rings, glass beads or baked clay [28,29]. Low biodegradable biomass such as straw and wood chips has also been found to be suitable as microbial carrier [30]. Microorganisms exist not only in the spaces within the carriers, but also attached to its surface, hence, a high-density microbial population is retained within the reactor, thus allowing a biomass retention time longer than the HRT [31]. The material should be non-biodegradable. The structure of the fixed film matrix should also be mechanically stable. Materials should be easily available in the local market at a reasonable cost. Different materials like nylon sponges, PVC, clay pipes, etc. had been used as support medium for fixed film reactors [32]. Fixed film technique has been used commonly for substrates of very low solids content where filters of very large surface area are used. However, but the studies are scanty with substrates of high solids content like cowdung slurry.

Many criteria need to be considered for selection of suitable materials for long life of the fixed film matrix [33]. Fixed film reactor packed with sponge nylon as support performed well in terms of specific biogas production rate as compared to conventional reactors. The results showed good digester productivity as well as satisfactory sludge stabilization in fixed film digester [34]. Vartak et al. found the performance of polyester medium with its high porosity and surface to volume ratio to be best both at 37 and 10 °C. It also yielded the maximum reduction in volatile solids (VS) and COD at 37 °C [35]. Weiland et al. obtained 75% and 25% reduction in HRT and reactor volume, respectively using plastic support for anaerobic digestion of screened cattle excrement as compared to conventional system. It had high accumulation of biomass during the 2-year study and could be operated in a broad range of loadings with a constant COD removal efficiency and high process reliability [36]. Raju et al. observed a high biogas yield of 0.70 m^3/kg VS added in PVCP packed reactors (20-day HRT) and COD reduction was three times more as compared to conventional reactor [37]. Henry et al. achieved high OLRs and a considerable reduction in the HRT utilizing random oriented plastic supports. This single stage reactor with recycle had many advantages like easy operation, homogeneous distribution within the reactor, maximum agitation, low risk of clogging or foaming and ease of control of biological activity by monitoring sludge activity [38]. Sanchez et al. used PVC plastic pipes and ceramic Rashig rings and found that anaerobic fixed-bed reactor could work at a high OLR without clogging. The efficiency of more than 60% in VS reduction and 55% in COD reduction were obtained at HRTs as low as 6 days [39]. Raju et al. used burnt coconut shells and obtained a high biogas yield of 0.72 m^3/kg VS added (20 day HRT) while Ganesh et al. achieved an increase of about 40% in gas production by the addition of broken burnt bricks as carriers for immobilizing microbes [37,40]. They suggested that larger lumps of bricks might be used to avoid clogging in practice. Sorlini obtained highest biogas production (144.0 l/kg VS fed) in the digester with wood chips while production was almost nil in the digester with expanded clay [41]. The number of anaerobic cellulolytic bacteria was considerably lower in the bottom sediment of the wood chip digester than in that of the expanded clay digester, whereas the number of methanogens was significantly higher. On the other hand Lomas et al. found lab and bench scale digesters packed with straight vertical channels of potter's clay to present large biodegradation efficiency and allowed large organic loads compared with continuously stirred tank reactor [42].

Several researchers had reported the effects of the pore size of bed material and bed type on the methanogenic performance and community in fixed-bed reactors. Sanchez et al. obtained a predominant presence of filamentous methanogenic forms closely resembling Methanosaeta (Methanothrix) located on the outer layer and in the bacterial framework of the biofilm, when they used six different support materials (polyurethane, bentonite, diabase, diatomaceous earth, sepiolite and PVC) for digestion of domestic sludge. The enhancement of CH_4 production was rapid and occurred within 24 h of sludge incubation [43]. Meier et al. found that energy substrates stimulated the attachment of *P. aeruginosa* and *Citrobacter amalonaticus* significantly as compared to the fixation behaviour in basal medium without substrates. This mechanism may be important for retaining methanogenic biomass in anaerobic biofilms and thus could help to reduce the start-up period of biofilm digesters and to enhance methanogenesis [44]. Recently, preliminary work on this concept at pilot scale (400 l, HRT$^{1/4}$30 day) has been carried out by Rana and coworkers at IIT, Delhi using stone chips and iron mesh biofilters. For the entire year, the biogas production from the reactor with iron mesh was consistently higher (~17%) than that from the conventional reactor. However, certain difficulties such as clogging of the reactor and decay of iron mesh after 1 year were encountered during the study [45].

D. Dalis et al. used two different types of anaerobic digesters to evaluate the anaerobic digestion of total raw olive-oil wastewater [46]. The main purpose of the study was to evaluate the performance of an up-flow-type reactor working in series with a fixed-bed-type reactor. The pilot-plant system operated in the mesophilic range (35 + 1 °C) during approximately 390 days, and

with organic loading levels that ranged between 2.8 and 12.7 g COD/l day. In a series of seven consecutive experiments, for the first stage (up-flow digester) optimum values of specific biogas production rate stabilised at a value of 2.1-l/(litre digester day) with a very satisfactory COD reduction of 83% (with a volumetric load of 11 g COD/(l day)). For the second stage (fixed-bed digester), the biogas production rate stabilised at a value of 0.22 l/(litre digester day) with a COD reduction of 8% (with a volumetric load of 0.19 g COD/(l day)). According to the results reported in the literature for total raw wastewater, the performance of the applied system is amongst the best for biogas production, COD reduction and loading rate reported so far, especially for the up-flow digester. Phenols were greatly reduced during the anaerobic digestion process in both digesters, with a concentration reduction, which reached 75% in the up-flow digester; with the use of the second stage (fixed-bed reactor) a further reduction of 45% was obtained.

With the above encouraging results, we may suggest, the employment of the up-flow type digester as an economical and effective treatment for significantly reducing the organic load of total raw wastewater. More satisfactory results might be expected from the use of a fixed-bed-type digester connected in series with a previous one, as a second treatment stage.

A recent study by Tatara et al. showed that a packed-bed reactor with CFT was applicable under very high organic loading rates and short hydraulic retention times, even if it was compared with other processes using supporting materials, such as anaerobic upflow fixed-film reactor or fluidized bed reactor. CFT provide the microorganisms with a place to grow and allow them to exist stably in the reactor [47]. Therefore, the microorganisms adhering to CFT are of great interest.

In a study, archaeal communities were compared in a packed-bed reactor operated on artificial garbage slurry (AGS), while the OLR was increased stepwise and the acetate-removing methanogen, *Methanosarcina* sp., became dominant among methanogens in the adhering fraction at an OLR of 12.2 g/(l day) [48]. However, the microbial population and its spatial structure in the adhering biomass in packed-bed reactors degrading organic solid waste have not yet been clarified.

Packed-bed reactors have been widely used for the treatment of wastewater. This can be well summarized in Table 1.

Anaerobic treatment of sugarcane molasses-based distillery spentwash using granular activated carbon (GAC) as support media showed an HRT of 4 days, corresponding to an OLR of 21.3 kg COD m^{-3} day^{-1}, with a COD reduction of 67.1% and gas yield of 0.45 m^3 kg^{-1} COD removed with 70% methane content [55].

The operational studies of a Pilot Scale Down flow Stationary Fix Film Anaerobic Reactor treating sieved piggery slurries (1 mm mesh) were presented using hydraulic retention time between 6 and 0.9 days. A medium organic load reduction was achieved, about 60% Total COD and 50% VS, both related to HRT longer than 3 days. Biogas productivity resulted in 4.5-m^3/m^3 day^{-1} for that HRT [56]. A pilot scale anaerobic fixed-bed process containing a high biomass concentration was studied. The fixed bed was filled with 25 Cloisonyle1 tubes (specific area: 180 m^2/m^3) and the support surface equaled 135 m^2. The support volume was 0.034 m^3, leaving 0.948 m^3 of effective volume [58]. The study showed that stillage of Sugar Cane Molasses is amenable to treatment by a fast anaerobic digestion process. Reactor was constructed in glass and filled with Rashig rings, the support material had an specific area of 11 cm^2/cm^3 [59].

When compared, both UASB and APBR showed greater than 90% COD removal efficiencies. An organic loading rate was increased from 1.5 to 7.0 g COD/(1 day) and the hydraulic retention times ranged from 13.2 to 2.8 days for both reactors. The methane yield increased with increasing organic loading rate up to 0.23 l CH_4/g

Table 1
Performance of packed-bed reactor for various wastewater

S. No.	Substrate used	Packing material	Specification of packing used	HRT (days)	Temperature range	Methane content (%)	Reduction in BOD/COD	Conclusion	Reference
1	Cellulose	Carbon Felt	Porosity# 92.2%, Sp.surface area# 0.70 m^2/g, Sp Wt # 0.11 g/cm^3	NS	Mesophilic and thermophilic	Mesophilic, 96.7; thermophilic, 49.2 (ml/g total solid fed)	NS	Carbon Felt has high porosity, rough surface and low bed density	[49]
2	Synthetic wastewater	Polyurethane foam matrices	NS	7 h	30 ± 1 °C	86–95%	NS	NS	[50]
3	Cheese whey	Charcoal, gravel, brick pieces, PVC pieces and pumic stones	NS	2	37 °C	72	Charcoal 81%	Charcoal bed proved to be the best supporting media for dairy waste	[51]
4	Palm oil, mill effluent	Pall rings	Diameter and height # 16 mm, surface area # 341 m^2/m^3	NS	NS	NS	90% during the first 12 days, thereafter, a decrease of 82.4%	The yield reached its highest level at 0.34 when Q_F (feed flow rate) and V_{up}(up flow velocity) were 1.01 l/day and 0.2 m/h, respectively	[52]
5	Olive mill wastewaters	Granular activated carbon	NS	NS	NS	NS	NS	*Proteobacteria,Flexibacter-Cytophaga-Bacteroides*, and sulphate-reducing bacteria present	[53]
6	Wastewater from a bulk drug industry	PVC pall rings	35 mm × 35 mm specific surface area 224 m^2/m^3	1.7 days	35 ± 2 °C	65–70%	COD # 47–66%; BOD # 83–86%	Efficient performance of the reactor up to an OLR of 10 kg COD/(m^3 day) is demonstrated	[54]
7	Cassava starch wastewater	Nylon fiber	NS	5.4	NS	NS	R_1 87%; R_2 84%; R_3 70%	The lowest number of methanogens in R_3 which correlated well to its relatively low efficiency	[57]

* NS, not specified.

COD degraded in the UASB reactor and 0.16 l CH_4/g COD degraded in the APB reactor [60].

Based on microscopic observation, the adhering biomass consisted of microorganisms, organic material, and void areas when an anaerobic packed-bed reactor using carbon fiber textiles (CFT) was operated using artificial garbage slurry. *Bacteria and Archaea* detected by fluorescence in situ hybridization was distributed from the surface to the inner regions of the adhering biomass. *Methanosarcina* sp. tended to be more abundant in the inner part of the adhering biomass than at the surface [61].

Diphasic fermentation of water hyacinth or garbage to biogas was tried by coupling a solid-phase acidogenic system (TS 20–30%) to an upflow, anaerobic packed-bed, methanogenic digester. In the case of fresh- and dried-water hyacinth significant levels of gas production occurred from the decomposing biomass bed and phase separation could be achieved only for short periods (25 days). Also, results indicated that the biomass bed may be used alone as a packed-bed digester to obtain high biogas production rates [62].

Three bench-scale bioreactors were operated to determine the effect of packing material and fungal predation on toluene capacities above $100\,\mathrm{g\,m^{-3}\,h^{-1}}$ were obtained in the fungal bioreactors packed with light-weight, artificial medium, and submersion of the packing in mineral medium once per week was found to provide sufficient moisture and nutrients to the biofilm. It was noticed that Polyurethane foam has a very open network of pores and, therefore, a large surface area. The fungus was found to grow in these internal pores rather than in the pores between the foam cubes. With the perlite, however, the fungus had a tendency to grow in the pores between the packing granules. This would account for the difference in pressure drop between the two systems, with a lower pressure drop being observed across the foam packing material [63]. A volatile solid analysis has revealed that about 75% of the biomass is attached to the inert media surface leading to a major contribution in the performance of the methane reactor [64]. High removal rates with 100% efficiencies of DMP removal were achieved up to the phthalate-loading rate of 560 g/m^3 h in a polypropylene tank (packed volume, 82 l) with 2.5 cm polypropylene open spherical packing units with a specific surface area of 7.9 m^2 and a porosity of 0.80 [65].

5. Comparisons of conventional reactor and APBR

Although there has been a great deal of research concerning the performance of high-rate reactors, there are few reports of comparative experiments between various high-rate processes treating potato waste leach ate operated under similar conditions [66].

The study was carried out to compare the performance of two types of bioreactors when treating high-strength leachate extracted from potato at various OLRs. The same leachate was treated in both a small-scale UASB reactor and an APB reactor with the overall aim of developing and comparing these [67]. The results served as the basis for alternative digester design such as two-stage processes. The high organic content and degradability of potato waste make it one potential source of renewable energy from agricultural waste in southern Sweden [68].

A valid comparison between data sets can only be made using experiments where the same waste and reactors of comparable size, and the same operating conditions are used. The methane yield, defined as the amount of methane produced for a given quantity of organic matter removed as a result of the activity of the anaerobic microorganisms [69], obtained from the APB reactor was similar to that reported by Mshandete et al. [66] during a study of the influence of recirculation flow rate on the performance of anaerobic packed-bed-rectors treating potato-waste leachate. Kalyuzhnyi et al. [70] obtained a methane yield of 0.3 l CH_4/g COD degraded at an OLR of 6 g COD/(l day) in a UASB treating potato–maize wastewater. In this study, the methane yields of both reactors were below the expected values of 0.35 l CH_4/g COD degraded [69].

However, despite its many advantages, anaerobic digestion is not widespread in the dairy industry, largely due to the problems of slow reaction, which requires longer hydraulic retention times and poor process stability using a conventional reactor. In order to solve these problems and to develop a better methanogenic process, several configurations of high-rate anaerobic reactors have been developed for treating soluble wastewater at relatively short HRT [71,72]. Retention of the active biomass independent of waste flow is achieved in advanced reactors [72,73] such as a fixed film reactor [72,74] which allows efficient digestion of high and low strength (in terms of suspended solids and organic materials) soluble wastes at much shorter HRT. The tied film reactor is capable of retaining active biomass in the reactor without the need for biomass recirculation. This capability becomes crucial under high organic loading conditions, because it allows the fixed film reactor to function efficiently without cell washout problem even at very high loading rates [71,74]. As a support for the growth of microorganisms, materials such as activated carbon and pumice stone have been reported [75,76]. However, the employment of anaerobic upflow fixed film reactor technology needs in depth analysis of biofilm formation and its performance during the steady-state conditions. There is a need to find means of improving the anaerobic digestion of potato waste leachate and increasing the methane yield. One possible way of achieving this could be codigestion with waste with a high carbon content to improve the C/N ratio.

6. Conclusion

A critical analysis of literature reveals that there is a strong possibility and need to enhance the use of anaerobic fixed film biogas reactors. This technique has many advantages over other conventional methods. Different materials (PVC, Pall Rings Polyurethane Foam, Carbon Felt, nylon fibers, straw, wood chips) had been tried as packing material depending upon their availability and other specifications. These packing materials would help to reduce hydraulic retention time and ultimately cost could be reduced. Practical aspects of using pure microbial film as magnifying microbial layers should be looked into.

It was concluded that the pore sizes of the packing material influenced methane yield. The increase in biogas production was explained by the increase in the quantity of methanogens immobilized on the packing material. Plastic support medium like PVC pipes were found to be more effective in reducing HRT.

Temperature played a great role on the yield and percentage of methane in Biogas from any substrate. Mesophilic temperature range proved to be better in both the context. This technique has been successfully tried for various substrates but the most promising one is the treatment of wastewater. There is a need to enhance anaerobic digestion using packed-bed reactors, for various other wastes too.

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